Master's Thesis in Materials Chemistry

# Recycling methods for glass fibre-reinforced plastic composites

Glass Fibre Composites in a Circular Economy?

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# Abstract

The reinforced composite industry is expanding. This raises the need for more sustainable resource use and waste management. A concept to combat both is circularity, creating a loop between the raw resources and the waste. Two important parts of circularity are recycling and reusing. This thesis presents literature studies on recycling, reuse and end-of-life methods for glass fibre-reinforced plastic composites, specifically with a thermoplastic matrix. The focus lies on preserving the reinforcement properties and either recovering the plastic matrix or its energy. Several viable methods were found, divided into thermal, chemical, mechanical, and other. The majority of the processes for thermoplastics can preserve the reinforcement properties while re-melting the polymer and reusing it. The main problem still is the novelty of the techniques and how to introduce them efficiently to the current composite supply chain. There is also a wider range of recycling techniques for post-industrial waste, posing the concern for post-consumer waste. Routes that can treat contaminated materials while still recovering their reinforcement properties are a subject for future research. There are also gaps in how to make it economically feasible and compete with inexpensive virgin materials. Despite the obstacles ahead, this thesis concludes that there are several potential routes on its way to becoming commercialised and this research field has just started uncovering knowledge to make the life of composites more circular.

# Är det inte dags att börja återvinna glasfiberplast?

**Ett växande miljöproblem är glasfiberkompositavfall. Idag existerar ingen tydlig återvinningsmetod utan istället läggs avfallet på hög eller förbränns. En cirkulär process som återvinner avfallet och introducerar det tillbaka in i tillverkningen kan gynna industrin både ekonomiskt och ur ett miljöperspektiv. Vad är dagens tekniska utmaningar och hinder som står i vägen för cirkulära kompositer?**

Dagens linjära levnadssätt, där produkter tillverkas, används och slängs, är inte längre hållbart och har aldrig varit det. Det måste förändras. Det måste bli cirkulärt. Cirkulär ekonomi är ett ord som ökat i popularitet i dagens samhälle de senaste åren. Cirkuläritet står för effektivare användning av planetens resurser där avfall återanvänds och introduceras tillbaka in i produktionskedjan. Om produkter från början designas utifrån ett cirkulärt perspektiv, kan både resursåtgång, utsläpp och avfall från materialindustrin minska. Cirkuläritet prioriterar material med lång livslängd och möjligheten att återvinna, utan att förlora produktens funktion. Ett exempel på en produkt med hög hållfasthet och lång livslängd idag är glasfiberförstärkta plastkompositer (GFPK). Kompositindustrin omsätter idag 1007 miljarder kronor och förväntas öka med 12% till 2027. En komposit är ett material bestående av två kemiskt olika material, designat för att utnyttja varandras styrkor och därmed skapa ett helt nytt material med andra egenskaper. GFPK består av fibrer av tunt glas inneslutet i ett plasthölje. Glasfibrer används för deras hållfasthet och kan absorbera tunga vikter utan att brista. Eftersom kompositen består av glas och plast är det ett rostfritt alternativ med låg vikt. De vanligaste användningsområdena för GFPK är i byggkonstruktion och bilindustrin samt i frätande miljöer och rymdfarkoster. En extra kategori är kommersiella produkter, hos företag som till exempel IKEA. Tillsammans med IKEA of Sweden AB skapades målet med det här examensarbetet: Identifiera på vilka sätt GFPK kan återvinnas. Som tidigare nämnts är GFPK redan designade för att ha en lång livslängd. Men i en cirkulär ekonomi måste GFPK också vara återvinningsbara eller kunna återföras till sin ursprungliga form.

På grund av materialets kombination av två fysiskt olika material och dess hållfasta natur, har det visat sig komplicerat att skapa en hållbar återvinningsprocess. Det här arbetet hade därför som mål att genom litteraturstudier undersöka och redogöra för vilka återvinningsmetoder som finns idag och hur de påverkar materialets funktion. Metoderna som undersöktes kan delas upp i fyra kategorier; termisk, kemisk, mekanisk och övrig återvinning. Trots den allmänna uppfattningen att kompositer är svåra att återvinna, observerades flera exempel på återvinning. Slutsatserna baserades sedan på hur användbara och effektiva metoderna har potential att vara om de skulle kommersialiseras. Termisk återvinning av GFPK innebär metoder som använder bland annat höga temperaturer för att förbränna materialet. Metoderna kan återvinna både energi och fibrer, även om avfallet var kontaminerat. En nackdel var dock att fibrernas mekaniska egenskaper försämrades av de höga temperaturer som användes. Ett intressant exempel på kemisk återvinning kunde återvinna både plasthölje och fibrer. Den kemiska reaktionen som användes hade förmågan att föra tillbaka plasthöljet till sina enklare beståndsdelar, vilket öppnar upp en möjlighet för plasten att återanvändas i en helt annan typ av produktionskedja. Mekanisk återvinning innebär istället att kompositen på olika sätt malas sönder och blir till små fragment av plast och fibrer. Fragmenten kan sedan återanvändas som utfyllnad i material som behöver förstärkning, men förlorar en andel av sin ursprungliga hållfasthet. Det här återvunna materialet kan vara lovande för industrier med något lägre hållfasthetskrav, som till exempel textilindustrin.

Fler lovande återvinningstekniker upptäcktes för plaster som kategoriseras som termoplaster. Detta innebär att de kan smältas ner och formas om, till skillnad från härdplaster som när de väl antagit en form och svalnat, inte kan smältas igen. Idag består kompositmarknaden av både härdplaster och termoplaster, där majoriteten är den förstnämnda. Men tack vare termoplasternas återsmältningsförmåga blir de alltmer populära. Flera metoder visade lovande resultat för GFPK av termoplaster där avfall och produktionsrester återvanns genom extra steg av smältning och omformning av kompositen. Fibrernas mekaniska egenskaper var i stort sett oförändrade och kunde användas för samma syfte även efter återvinning. En utmaning för dessa metoder är att inkludera kontaminerade rester eller komsumentavfall i återvinningsprocessen. Färgning, utfyllnad och tillsatser är delar av materialet man helst inte vill ha med i en återvinningsprocess. Det är också svårt att konkurrera med den billiga tillverkningen och det låga pris som dominerar dagens glasfiberplast. Trots detta är det lika viktigt att inte avfärda kompositernas cirkulära livslängd, utan istället dedikera mer forskning för att lösa problemet i en marknad som bara växer. Nya metoder som efterbehandling av fibrerna för att återfå deras mekaniska egenskaper efter återvinning möjliggör ett nytt sätt att se på produkttillverkning. Om processerna utformas för cirkularitet från början blir övergången mycket lättare att kommersialisera. Forskningen för cirkulära kompositer har bara börjat, frågorna kring återvinning kvarstår. Låt oss göra mer och se GFPK bli cirkulära i framtiden.

# Acknowledgements

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# 1

# Introduction

## 1.1 Background

Composite materials are growing in popularity in market segments such as construction, the automotive industry, corrosive environments, commercial products, aircraft, and spacecraft applications [16, 20]. The material has been designed with the intention of often replacing heavier materials using less mass, but still having the same strength properties. Some examples of commercial composites are reinforced concrete, plywood, fibreglass, reinforced polymers, metal matrix composites, and ceramic composites. Some other examples of benefits of composites are, but are not limited to: Insulating, non-magnetic, high strength-to-weight ratio, corrosion-resistant, radar transparent, low maintenance and high impact strength [4]. Today's market for GFRP composites had a revenue in 2020 of $99.91 billion, it is predicted to increase by 12% by 2027 (Insights, 2019). The polymer matrix segment is the leading one, compared to the other two common composite materials that use metal or ceramic matrices. This will inevitably lead to increased manufacturing waste and a need for sustainable and circular end-of-life solutions.

The GFRP is rarely recycled today due to their heterogeneous composition, long life span, cheap virgin materials and, for some methods, tensile strength loss after recycling (Yang et al., 2012). From a long term perspective, recycling or reusability is crucial for materials to still be viable. This thesis will investigate recycling and end-of-life solutions to give a perspective on whether GFRPs can be included in a future circular economy. Introducing the terms *circularity* and *circular* economy. The concept of circularity for materials means developing products meant to last, that are easily reused and recycled at their end-of-life without losing their functional value [3]. Circularity promotes making better use of all resources, fully recovering waste and thus closing the loop of resource flow. A circular economy stands for preventing further pollution by designing products with higher quality that can be used for a longer time. One example of such a product is glass fibre-reinforced polymer composites, or in short GFRP, built to last longer, with the use of less material than its unreinforced polymer counterparts.

## 1.2 Research Problem

This thesis will investigate glass fibre-reinforced thermoplastic waste and what impact recycling has on the composites' material properties. Collaborating with IKEA of Sweden AB, an interested party in composite recycling, the focus will be on researching recycling methods and property changes for glass fibre-reinforced thermoplastics, as well as some comparison to thermoset matrices. The composite market today is dominated by thermoset resins but the trend to use thermoplastics instead is slowly catching on (Cousins et al., 2019). Thermoplastics can be re-moulded and re-heated after use and require lower processing temperatures. These characteristics make thermoplastics more interesting from a circularity aspect, and will therefore be the main focus of this thesis. The research aims to increase the knowledge and awareness of composites recycling routes for interested parties, for example, glass fibre suppliers, reinforcement processing, construction and others in the industry. If the stated recycling methods show enough promising results in maintaining mechanical properties then the knowledge can be beneficial to mentioned parties. By further reducing the gap to closing the loop on glass fibre processing, it is possible to recover both energy and materials.

**Research questions**

- What are some ways in which GFRP can be recycled or reused?
- What happens to the properties of the GFRP materials after different kinds of recycling?
- Which recycling or end-of-life methods are the most viable for GFRP composites?

## 1.3 Aim and Goal

This thesis aims to give an overview of today's GFRP recycling methods and their changes to the recycled material properties. The goal of this thesis is to identify, compare, and present end-of-life applications and recycling methods of the GFRP composite. Additionally, this thesis will identify and present if and how the methods change the material characteristics after recycling, specifically the reinforcement and adhesion properties.

## 1.4 Contribution of knowledge

The purpose and knowledge contribution of this thesis is to give an updated picture of recycling GFRPs. Research and summarising literature studies will increase the knowledge of relevant recycling processes and changes to material properties. A greater understanding of possible composite recycling routes will narrow the gap to circularity.

## 1.5 Constraints

The thesis will not discuss the composite recycling industry concerning direct cost, production, or capacity, but instead focus on the processing and material properties aspect. It will briefly mention other composites like carbon fibre-reinforced as a comparison, but this will not be the main focus. It will not dedicate a lot of focus on chemical recycling routes, since these often produce environmental concerning chemical waste, which is unfavourable in a circular economy. Neither will it discuss biological degrading processes like biological recycling, since fibre-reinforced composites are built to have a long life-span, and thus not being biodegradable by nature (KIS, 2020). The thesis will be based on literature studies from legitimate, peer-reviewed scientifically published articles. The main portion of the references has been gathered from well-established databases, including Research Gate [14] and ScienceDirect [12].

## 1.6 Outline of the thesis

This thesis will begin by defining what a glass fibre-reinforced composite is, mentioning sizing and glass types. After that, it will move on to exploring recycling and end-of-life solutions, dividing them into different categories: thermal, chemical, mechanical and other. These sections will also include reported changes to material properties. Lastly, the literature studies are summarised and compared in an analysis discussion. The thesis will finish with a conclusion on which methods are recommended and future recycling research.

# 2

# Glass fibre-reinforced composites

This thesis will use the following definition when referring to a composite: a composite, or composition material, consists of two or more constituent materials with different chemical or physical properties. The materials are either categorized as "matrix" or "reinforcement" designed to act as one, but remain separate as they never fully merge [4]. The idea is to create a material with different properties than the individual material components.

## 2.1 Fibre-reinforced plastic composites

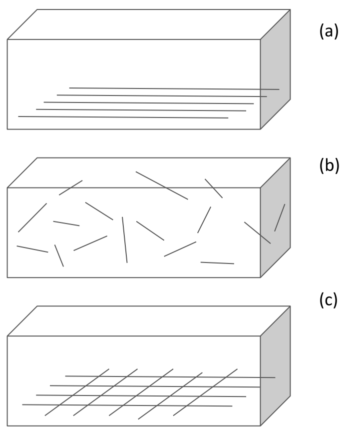


*Figure 2.1: Figures of different fibre orientation in a polymer matrix: a) 1D, b) randomized and c) 2D grid*

Glass fibre-reinforced polymers, or GFRP, consist of high-performance fibres embedded in a polymer matrix. The fibres can reinforce in different dimensions to give different properties, see Figure 2.1. The 1D and the grid orientation are the most common. The function of the fibres is to take the maximum load while the function of the polymer matrix is to transfer the stress between the fibres while at the same time protecting them from mechanical and environmental damage (Gettu, 2015) (Sinha, 2020). This is why adhesion between the fibres and the matrix is important.

### 2.1.1 Different types of glass fibres

The by far most common type of glass used as reinforcement fibres are called E-glass, short for electrical glass because of its electrically insulating properties. Its popularity is due to its low production cost and early development compared to other types. The E-type consists of alumino-borosilicate glass and less than 1% alkali-oxides. Borosilicate glass has a low thermal expansion coefficient and can withstand temperature changes without fracturing. The E-glass is also water-resistant because of the low alkali content but is easily destroyed by acids.

### 2.1.2 Sizing

*Size* is a thin coating, often consisting of organic material, applied to the surface of the glass fibres during manufacturing. Sizing is the process of coating the fibre. The sizing of the fibres is considered one of the most important parts of the manufacturing process to ensure favourable reinforcement properties (Thomason, 2019). The homogeneous, thin coating is applied to the surfaces of the fibres during processing for handling protection. When the fibres are in use, the sizing provides better adhesion between fibre and resin matrix, thermal stability and chemical resistance [22].

# 3

# Recycling and end-of-life processes

The current recycling routes for GFRP composites can be divided into four categories: thermal recycling, chemical recycling, mechanical recycling and others. Other in this report means the remaining methods that don't fit in the previous categories, for example, high-frequency recycling and injection moulding waste. The section consists of investigated and summarized literature studies of recycling methods and material changes. The sections will include when provided by sources if the methods are applicable for Post-Industrial Recycling(PIR) or Post-Consumer Recycling(PCR). PIR mainly refers to waste generated by the manufacturing process of the source material, in this case, the manufacturing of the composites [28]. PCR refers to finished products that have been used and later recycled. PCR is, therefore, more contaminated and often harder to recycle since it can require additional cleaning and separation steps.

It is important to note that the majority of the studies on recycling glass fibres have been performed on composites with thermoset matrices, developed to often only recycle the fibres and not the resin (Bernatas et al., 2021). Studied methods in this report will also look more into thermoplastic matrices, since that calls for recycling of both the matrix and the fibres, and align more with the idea of circularity. The studies will include all reported material changes but focus on the preservation of mechanical properties. Keeping the function of the material even after recycling can lead to it having a longer life span and getting a higher use-value of the raw materials.

## 3.1 Thermal recycling

Thermal recycling processes will have the material go through a high-temperature treatment, with the potential of recovering some solids and some energy (KIS, 2020).

### 3.1.1 Pyrolysis

The pyrolysis process typically operates within 450 and 600 degrees Celsius (Torres et al., 2000), and the method is considered an alternative to mechanical recycling for thermosets that cannot be remoulded. Pyrolysis is heating the material in an atmosphere without oxygen, to make it degrade without burning. The material turns into gas, oil and various solids(fibres, fillers and charcoal). A study from 2018 noticed that the fibres came out with a charcoal residue on them. Charcoal can be minimized by using a small amount of oxygen during the process (Naqvi et al., 2018). Another alternative is post-pyrolysis oxidation to burn off the charcoal residue from the surface of the fibres to get a clean finished product. Pyrolysis will therefore allow fibre and filler recovery, degrading the material without impacting the fibres form. The resin will break into low-weight molecules and produce gases and oils, which the mentioned study suggests using as fuel or chemical source of energy. The inorganic compounds, the fibres and the calcium carbonate, mechanically unmodified, have the potential to be recycled into other composites or plastic material. Since the process filters the solids and the resin, this method could be viable for PCR, depending on what type and what grade of contamination the used waste contains. Considering the harm to the fibre's mechanical properties, this may be economically challenging, to compete with the inexpensive production of virgin glass fibres.

While recycling using pyrolysis shows the most promising results for carbon fibres, it is not the same for glass fibres. The thermal degrading harm the tensile strength of the fibre, which is often one of the key components in using glass fibres as reinforcement. This due to the heightened temperature slightly impacting the fibres molecular configuration that may induce cracks. Several property changes to glass fibres from either direct temperature elevation or recycling have been studied. In almost all cases, a decrease in tensile strength. Despite several literature studies, the direct mechanisms behind this loss are still unclear, but probably related to the induced cracks.

### 3.1.2 Fluidised bed pyrolysis

This method starts with having the composite reduced in size by shredding and then passed through a fluidised bed of silica sand (Pickering et al., 2000). The material is heated between 450 to 550 degrees Celsius and the fibres are released by attrition as well as thermal degradation of the matrix. The resin will decompose into oxidized molecules and fibre filaments. These get separated by carrying lighter components up an airstream, while heavier ones sink to the bottom of the bed. This separation ties to the advantages of this process, that it can treat both contaminated and mixed materials, for example, coated surfaces, metal inserts and foam cores. The study concludes that this is a suitable option for end-of-life and post-consumer waste, though not yet commercialized. The disadvantage is that the fibres get damaged, keeping their stiffness but with a reduction in tensile strength. approximately 50 % reduction with a working temperature of 450 degrees. Compared to the pyrolysis process, the fluidized bed does not recover as much energy, since only gases are recovered(Pickering, 2006).

### 3.1.3 Microwave assisted pyrolysis

The theory behind microwave-assisted pyrolysis, or MA pyrolysis, is heating the materials core to enable fast thermal transfer, and therefore saving energy. The heated material absorbs the electromagnetic energy from the microwaves and converts it into thermal energy. Conventional heating will transfer heat from the surface to the inside, while this technique directly heats the material's inside (Bernatas et al., 2021). This technique is only applicable to microwave absorbing materials, not for example S glass or Teflon that are microwave transparent (BB, 2018). MA pyrolysis enables more moderate temperatures, which favours the preservation of the glass fibres' mechanical properties. The method degrades the resin matrix into basic components, such as monomers and fillers.

A study from 2012 investigated MA pyrolysis recycling GFRP from a wind turbine blade where the reclaimed fibres had 70% of the initial mass (Åkesson et al., 2012). However, the surfaces of the fibre were covered in char residue which leads to poor adhesion between fibres and the polymer matrix. But as mentioned before, the char can be minimized with the use of oxygen. The study suggests that an application could be using partly recycled fibres with virgin fibres, to produce a composite with slightly lower mechanical demands. A report from 2016 by Composites UK claims that this process has been studied and tested academically but has not yet been commercially successful (Job et al., 2016).

### 3.1.4 Chemical solvolysis

Solvolysis is a type of nucleophilic substitution where a heated, reactive solvent dissolves the resin matrix into low molecular weight chemicals (Cousins et al., 2019). The reaction breaks the covalent bonds of the polymer matrix, a process often requiring elevated temperatures and pressures. The glass fibres are not dissolved and will break free from the mix, however, the process degrades the fibres by removal of sizing, hence removing thermal stability and protection as mentioned in section 2.1.2. Companies have already prepared solvolysis technologies to sell and distribute. For example Adherent Technologies Incorporation in the USA, and more recently Innoveox in France as well as Panasonic Electric Works in Japan that have built a pilot plant for their water-based solvolysis process to recycle 200 tons of GFRP manufacture waste annually. The performance of the fibres' mechanical properties will depend upon the process conditions. Tests have shown that the glass fibres' tensile strength can be reduced from 52 to 64%, similar to pyrolysis. This drop highly depends on the temperature during the solvolysis. A higher temperature range of 500 to 550 degrees is often required to treat thermoset matrices. Unfortunately, this high temperature damages the fibres and strips them of their tensile strength, their important characteristic to be used as reinforcement (Thomason et al., 2016). This also results in harder processability. Most studies on solvolysis have however been made on the resin part of the composite and lesser on the glass fibres. There is a significant potential to reuse resin products, in the form of monomers or additives. But the process is still in need of research since it will also need purification and separation steps. It also potentially uses hazardous solvent chemicals, as well as expensive equipment to benefit the reactions. The conclusion from the mentioned studies suggests that chemical recycling is a lesser suitable option to recover the fibres in GFRPs and more beneficial for resin recovery, similar to pyrolysis recycling.

## 3.2 Mechanical Recycling

Mechanical recycling processes typically consist of shredding the composite waste and separating fibre-rich and matrix-rich sections. These materials are usually lower quality compared to before (KIS, 2020).

### 3.2.1 Milling

One way of mechanically recycling GFRP is first reducing the size by breaking up the material into smaller flakes and then grinding it in a hammer mill (Job et al., 2016). The method is discussed in the same study as mentioned before in section 3.1.3, the 2016 report by Composites UK. The different sizes can be separated and used for different things. The recyclates will probably consist of resin powders, fibres and material flakes. The fibres are degraded of their length and strength properties but the use of these ground composites has the potential to either be as a filler or reinforcement. However, as mentioned before, virgin based fillers are still much cheaper than recovered fillers. The amount of filler in a new material is limited due to deterioration of the mechanical properties and harder processing, the latter due to an overall higher viscosity. The breakpoint is less than 10wt%. There are already industrial applications utilizing this among GFRP manufacturers (García et al., 2014).

### 3.2.2 High Frequency Recycling

A brief mention of a new potential method of recycling is presented by the Swedish company Librixer AB. By using air flows to create high-frequency rotation of the material waste it forces the pieces of waste to bounce off the walls and break by impact damage. The Librixer machine will therefor break the chemical bonds of the material without the use of elevated temperature, water or chemicals [2]. A recent study performed at RISE indicated that the Librixer technology has the potential to separate thermoset matrix from glass fibres to a large extent [21]. The fibres and matrix were still physically blended and a method to separate the matrix particles from the glass fibres will be required to acquire glass fibre fractions with only minor matrix residues. The company claims to be able to recycle wind turbine blades and separate the fibres and the epoxy resin, but it is still in the start-up phase and is still investigating potential areas of usage.

### 3.2.3 Dissolution and Injection Molding

A promising route is reusing the thermoplastic GFRP waste in injection moulding after the process of dissolution. Dissolution is the process of breaking the chemical bonds of a polymer matrix. More specifically, it can be described as physically dissolving the bonds of the matrix in a solvent. Only for thermoplastic matrices, dissolution enables the recovery of both glass fibres and polymer matrix while maintaining the stiffness and strength of the fibres throughout the recycling process. A study shows that the injection moulded waste had equivalent mechanical properties or even stronger compared to virgin material (Cousins et al., 2019). This is likely due to thermal healing of age-induced cracks during the injection molding process. The technique also enables thermoplastic recycling using lower temperatures as it will recover the polymer matrix as well as the fibres. Conclusions from a 2019 study by the Colorado School of Mines showed that glass fibres, in a thermoplastic composite, had equal tensile strength and a 12% reduction in stiffness, after recycling by dissolution. The study also showed that the material recovery suffers some losses in the process, averaging the resin recovery to 90% and fibre recovery to 50%.

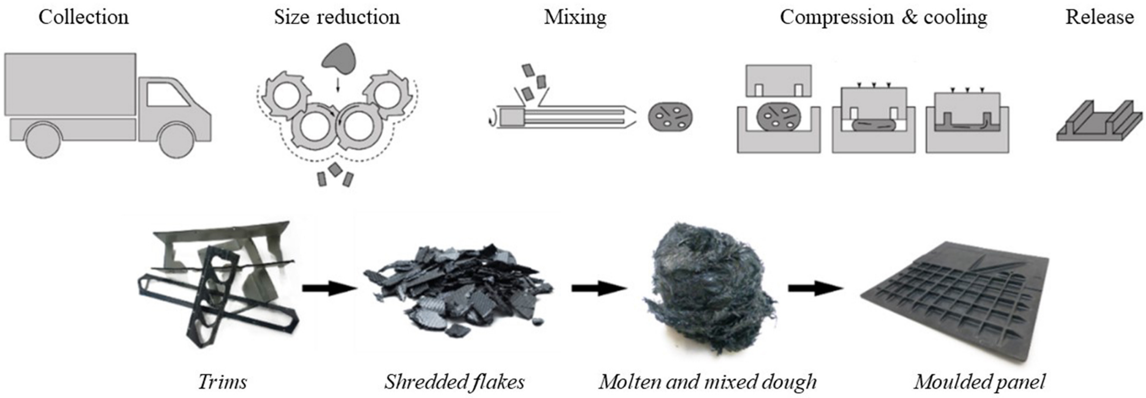


*Figure 3.1: The TPC-cycle recycling process [8]*

## 3.3 Other recycling methods

### 3.3.1 TPC-cycle Recycling Project

This method is a project by the ThermoPlastic Composites(TPC) Application Centre in the Netherlands, partnering with the industry to target production scraps and developing a new recycling route [8]. The goals are to preserve mechanical properties and at the same time reduce the environmental impact, without making it at an unreasonable cost. Their solution is said to produce net-shapes manufacturing, which are 3D-shapes that are opened and laid flat, as well as complex forms in a short time by recovering both the fibres and the matrix. The process starts by size reducing the composite to flakes and then heating and shear-mixing simultaneously the materials into a dough (de Bruijn Thomas and van Hattum Ferrie, 2020). The dough is compression moulded in an isothermal mould and the final product gets released as a panel, see Figure 3.1 above. The trims are collected from production scraps and assumed to be directly used in the process, which means that fillers and different colouring will remain and be present in the final product. The retention of the long fibres enables the high preservation of mechanical properties. A PhD thesis, by Guillaume Vincent in 2019, focused on the relation between the wanted material properties and the processing steps (Vincent, 2019). It aimed to develop workability and economic viability for multi-layered woven materials. The conclusions from the technical point of view were that the polymer resin was degraded with a change in crystallinity. Another study built rotorcraft panels from TPC recycled carbon polyphenylene sulfide). The panels with recycled material proved to be lighter, more cost-effective and approved when flights were tested (de Bruijn Thomas and van Hattum Ferrie, 2020).

### 3.3.2 Thermosaic and Thermoprime

The Thermosaic® and Thermoprime® commercially available techniques are developed by CETIM CERMAT, a research centre in Alsace, France [9]. The processes are specifically developed to produce thermoplastic composites from recycled thermoplastic. Thermosaic uses thermoplastic composite manufacturing scraps in a thermo-mechanical process. The first step is a fragmentation of the waste material, followed by re-agglomeration. The treated material is then milled and again broken into small pieces that are laid on a conveyor belt to form a homogeneous bed. The bed is then heated and compressed to produce panels up to 10 mm in thickness. The mechanical properties of the fibres are preserved due to the conservation of fibre length. The original length can be a maximum of 200 mm. Thermoprime has a slight change in input, where it upcycles recycled thermoplastic, for example, PP or PA, with long-length fibres. This technique results in composites with better properties than before because the process compensates for the loss caused by ageing and the previous recycling step. Compared to previously mentioned recycling methods, this one introduces a possibility to the production of virgin GFRP manufacturing, using recycled thermoplastic instead of virgin. The systems both produce panel-shaped products that can be further processed by thermoforming or thermocompression.

## 3.4 End of life applications

The following section will present a few ends of life applications who do not require any recycling.

### 3.4.1 GFRP in Cement Kilns

Previous studies have shown that GFRP has a promising end-of-life application where it gets co-processed with refuse-derived fuel in cement kilns. Refuse derived fuel is fuel from various waste processes, industrial and commercial, replacing traditional fuels like coal or fossil. In 2011, The European Composites Industry Association recommended that GFRP waste is recycled this way. Their report, which investigated chemical, mechanical and co-processed recycling, stated that the co-process in the cement kiln will utilize both the energy and the materials, as will be explained further in the section. In comparison, the report states that mechanical and chemical only as of now recycle the material (Backer, 2011).

GFRP typically contains E-glass, as mentioned in section 2.1.1, the alkali-free alumino-borosilicate, in a matrix of an organic resin, sometimes with a calcium carbonate filler. This is beneficial for the cement kiln because the resin burns off providing energy and the mineral constituents provide feed-stock for the clinker, the ground to form cement. Calcium carbonate is a primary component of Portland cement, which is the most common cement type, as well as alumina and silica. Boron can however cause a reduction in strength, but only in the setting of the cement, and low proportions are not considered a problem. E-glass from Europe contains lower and lower amounts of boron due to emissions regulations, compared to higher amounts in E-glass from China. The study concludes that introducing GFRP waste to be used in cement kilns has two major advantages compared to just burning the material for energy. The mineral content is recycled into the cement clinker, compared to burning into ash, and the energy provided by the resin will lower the demand for fossil fuels in the process. A study from 2011 states that this recycling method results in 67% material recovery and 33% energy recovery (Jacob, 2011). The method is an immediate solution that is capable of treating big volumes without producing ultimate waste.

## 3.5 Post-treatment of GFRP

Previous recycling examples show ways of recovering glass fibres, some with the trade-off of losing their strength and stiffness. A post-treatment to recover these properties of recycled glass fibres has been suggested by the University of Strathclyde in Scotland, in collaboration with the wind power company Aker Offshore Wind. The project sparked from the increasing amount of GFRP wind turbine blades that get discarded in landfills or turned into waste energy. The project claims to achieve near virgin fibre properties by a thermal post-treatment after recycling to restore tensile strength [25]. The patented method states recovery of up to 80% compared to virgin fibres. It further claims to have the potential to manufacture fibres at a lower cost and introduce them to the existing supply chain [24].

## 3.6 Environmental impact of recycling methods for composites

The environmental impact of composite recycling is highly dominated by energy usage. The majority of the manufacturing and recycling processes have the electrical energy demand as their biggest footprint. Therefore, the big goal is to reduce energy usage in recycling composites (Job et al., 2016).

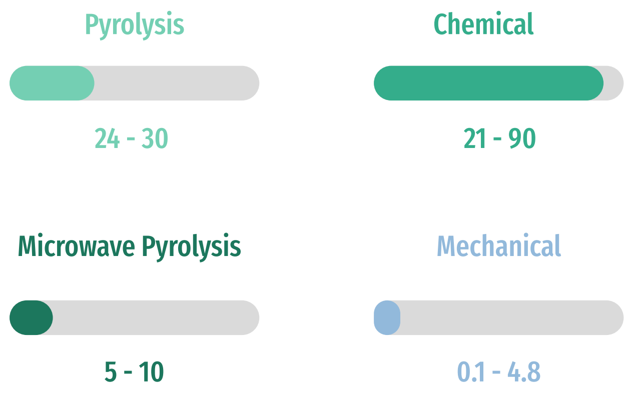


*Figure 3.2: Energy demand in composite recycling methods [MJ/kg] (Job et al., 2016).*

As Figure 3.2 above shows, chemical processes typically demands higher energy, compared to others. But this can be off-setted by the potential to gain value from the fibres and resin chemicals. Pyrolysis techniques use less energy but damage the fibres' mechanical properties. Mechanical grinding uses even less energy, but also produces a lower value product (Backer, 2011). The Figure remade with higher resolution, comes from a 2016 study on composites recycling done by Composites UK, mentioned before in 3.4.1. The energy values for mechanical recycling will vary depending on what machine types are used, what capacity they have and how much it operates at full capacity, which is where the energy demand is at its minimum. The study also wants to note that some processes can generate energy and materials. As an example, the cement kiln route incinerates the GFRP waste for energy recovery while the incombustible parts are used as raw materials in the cement industry. Lower environmental impact can be received through avoiding virgin materials and introducing recyclates in the current production routes. Composites UK highlights that limiting new production of high embodied energy materials, such as fibres and polymer materials, will also gain environmental benefits. Integrating with existing waste management and production supply chains are two important steps to enable commercialisation. Another more recent study from 2021 concludes that more research is still needed in the following areas: designing composites to be recyclable, more efficient recycling processes, production techniques using recycled materials and improving the waste collection chain (Bernatas et al., 2021). The market also needs to work toward more cost-effective processes, since it is now less expensive to dispose of composites in landfills, than reusing or recycling, even feeding it into cement kilns. The authors of the study also state that the most advanced recycling techniques today are mechanical grinding for glass fibre-reinforced polymers and fluidized bed pyrolysis for end-of-life and contaminated GFRPs. However, it also highlighted that there is potential for new thermoplastic composite routes because of their significantly easier recyclability compared to thermosets, like the Thermosaic and Injection Molding Waste processes mentioned before.

# 4

# Analysis

## 4.1 Results

The literature studies concluded several recycling methods for glass fibre-reinforced polymers, summarizing key points and comparing them in the figures below. The original tables are found in Appendix A. The analysis behind the results is discussed below in section 4.2.

| ***Studied Methods*** | ***Resin rec.*** | ***Fibre rec.*** | ***Energy rec.*** | ***Fibre changes*** | ***Advantages*** | ***P*** | ***Drawbacks*** |
|---|---|---|---|---|---|---|---|
| Pyrolysis | No | Yes | Yes | Damage fibres | - Filler recovery<br>- Can use the produced gases and oils as fuel | / | - Char contamination on components<br>- Produces gas emissions<br>- Fibers degraded by high temperatures |
| Fluidized bed pyrolysis | No | Yes | Yes | Damage fibres | - Can treat contaminated and mixed materials<br>- No char residue | PCR | - Loss of tensile strength on fibres<br>- Lesser energy recovery compared to pyrolysis |
| Microwave assisted pyrolysis | No | Yes | Yes | Damage fibres | - Less harm to fibres because of lower temperature<br>- Degrades resin into monomers | / | - Char residue on fibres<br>- Only for microwave absorbing materials |
| Solvolysis | Yes | Yes | No | Loses sizing | - Resins return to monomer state and can easily be reused<br>- No char residue | / | - Expensive equipment<br>- High temperature and pressure conditions<br>- Possible hazardous solvent |
| Milling | Yes | Yes | No | Damage fibres | - Milled material can be used as filler<br>- Low energy cost | PCR | - Loss of mechanical properties for fibers, both length reduction and degradation<br>- Filler usage is limited by hard processability due to high viscosity |

*Figure 4.1: Comparison chart part 1.1 of studied recycling methods. The empty slots indicate that it is either not relevant or missing information (P stands for Post, meaning Post-Industrial or Post-Consumer and Rec. stands for Recovery). The backslash indicates that it can be both*

| Studied Methods | Resin rec. | Fibre rec. | Energy rec. | Fibre changes | Advantages | P | Drawbacks |
|---|---|---|---|---|---|---|---|
| High frequency | Yes | Yes | No | Poor adhesion | - Uses no chemicals, heating or water<br>- Breaks the weakest chemical bonds | PCR | - Start-up stage<br>- Need separation stage and cleaning off the polymer residue from the fibers |
| Thermosaic & Thermoprime | Yes | Yes | No | Stiffness reduc. | - Preserves mechanical properties. | PIR | - Only for thermoplastic manufacturing waste |
| ThermoPlastic Composite-Cycle Recycling | Yes | Yes | No | None | - Preserves mechanical properties.<br>- Retention of long fibres. | PIR | - One study show a slight degrading of the matrix, and a change in crystallinity. |
| Injection Molding Termoplastic Waste | Yes | Yes | No | - | - Equivalent mechanical properties for recycled fibres compared to virgin | / | - Only for thermoplastics<br>- Dissolution step requires volatile solvent<br>- Material recovery loss |
| Cement Kiln | No | No | Yes | - | - Can handle big volumes with no ultimate waste<br>- Utilize both material and energy | PCR | - One study reported only 67% material recovery and 33% energy recovery |

*Figure 4.2: Comparing chart part 1.2 of studied recycling methods.*

| Studied Methods | Thermoplastics (TP) / Thermosets (TS) | Country | Commercially available | Associated companies |
|---|---|---|---|---|
| Pyrolysis | TS | Europe | Yes | - |
| Fluidized bed pyrolysis | TS | - | No | - |
| Microwave assisted pyrolysis | TS | - | No | - |
| Solvolysis | TS | Japan, USA | Yes | Innoveox<br>Adherent Technologies Inc<br>Panasonic Electric Works |
| Milling | TS / TP | - | Yes | - |
| High frequency recycling | TS / TP | Sweden | No | Librixer AB |
| Thermosaic & Thermoprime | TP | France | Yes | Cetim Cermat |
| ThermoPlastic Composite Cycle Recycling | TP | Netherlands | No | ThermoPlastic Composite Application Centre |
| Injection Molding Thermoplastic Waste | TP | - | - | - |
| Cement Kiln | TS | Germany, USA | Yes | Eco Wolf<br>ADM Isobloc |

*Figure 4.3: Comparing chart part 2 of studied recycling methods.*

## 4.2 Discussion

The studied methods were interpreted from a technical point of view, ranking high mechanical property preservation as the number one criterion since property preservation enables recycling or reusing the material for the same purpose as before, thus giving it a longer lifespan. To tie back to the introduction, this will minimize the need for virgin composite manufacturing and contribute more to the concept of circularity.

Of all the thermal recycling methods, pyrolysis may be the most researched and commercially tested. However, the individual conclusions are that microwave-assisted pyrolysis does the least damage to the fibres' mechanical properties since the process temperature is significantly lower. But the method still introduces too much fibre damage to be considered from a circular perspective. The fluidized bed opens the possibility to recycle contaminated materials with fewer processing steps and lower fibre requirements. The separation step where the heavier materials sink to the bottom of the bed and the lighter materials travels upwards with the air stream also removes a step in the process. Several other mentioned methods chemically or mechanically separate the two materials but they still exit the recycling together, which is why the introduction of these recycling methods will require a separation step to be useful.

From a first glance, the Librixer recycling process seems more than just promising. It is a mechanical method, which means less energy usage, as shown in 3.2. It does not use any chemicals or water and breaks the composite mechanically by its weakest bonds. As section 3.2.2 explain, recent tests revealed that the resin and the fibres exit the process in a pile, and the fibres need to be cleaned off of resin residue to be able to adhere to a new polymer matrix. It is not clear how else the process affects the fibers, like change in length for example. If these technical aspects can be solved, the high-frequency method is an interesting technology to keep an eye on. Since the company Librixer is still in the start-up phase, there can be a lot of development and changes in the coming years. Another new and novel technique, this time by Cetim, is Thermosaic and Thermoprime, specially designed for thermoplastic composite waste. The several steps processes produce panels with preserved mechanical properties, due to the conservation of fibre length, with lengths under a certain mark. These panels can later be as an example thermoformed to the desired shape. Thermoprime up-cycles already recycled thermoplastics with virgin fibres. Both techniques show promising results to introduce recycled materials in virgin GFRP manufacturing. Thermosaic can be adopted by the current manufacturing streams to minimize and reuse scraps, while Thermoprime can be an alternative to virgin thermoplastic matrix production. The techniques are commercially available and hopefully, Cetim keeps on developing them even more. A similar technique is the TPC cycle project by the Thermoplastic Composite Application Centre. The project was created to preserve mechanical properties and reduce environmental impact. Similar to the Thermosaic process, this too uses production scraps and produces it into a panel that can later be re-formed. The process of shear-mixing and heating was shown to slightly degrade the polymer and change its crystallinity. What this means for the properties as a composite, remains to be studied. As it is not commercially available, the project is still undergoing research and may propose changes to the steps and handling. This can enable better properties and make it more interesting to look into in the future.

The useful aspect of thermoplastics and their ability to perform dissolution was found to be utilized by injection moulding glass fibre-reinforced thermoplastic waste as investigated in section 3.2.3. A step to break up the waste is needed, to form it into granules or pellets to feed into the hopper. It was reported that the material's mechanical properties have negligible to no change at all after recycling. This is a route to recommend for especially post-industrial thermoplastic waste since all fillers and colours in the original products will probably remain. Compared to Thermosaic and TPC-cycle recycling, the injection molder require less steps in terms of processing. It is also already widely used today, and that may pose a possibility to ease the introduction of the technique in current supply chains.

The post-treatment 3.5 claimed to give an 80% strength recovery to recycled glass fibres. This proposes the potential to reuse them in similar reinforcement applications as they previously served. That poses other questions if the treatment can be introduced easily into the current supply chain if it affects the sizing of the fibres and therefore the adhesion properties to the polymer matrix. From a long term perspective, it can be important to investigate how many per cent will be recovered the second post-treatment. For the sake of circularity, this matters, but for the planet's current state, one post-treatment and renewed life span of the fibres can be considered good enough. If the treatment can be introduced into the current manufacturing stream, it has the potential to be more than good enough can start creating the loop of recycled materials in composites. If the mechanical properties partially degrade each loop, they can be used as fillers and still reduce the virgin materials used. The thermal and chemical recycling techniques may especially need some form of post-treatment, if recovering the fibres function is prioritized. This connects to 3.6 where the European Composites Industry Association encourage the composite field to find more applications for recycled glass fibre fillers. Introducing fillers in the current supply chain is another way of reducing virgin material usage. But as mentioned earlier, virgin glass fibre production is widely adopted and inexpensive, a hard competitor to recycled fibres. Fillers may also come from the mechanical recycling of milling 3.2.1. Milling produces a lower value product by physically breaking up the material, but uses a lot less energy to do so, compared to thermal and chemical recycling. It all comes down to summarising the overall environmental impact. However, in terms of circularity, milling does not provide a longer life span or a high yield of recovered material and is therefore not recommended for recycling GFRP.

Depending on what range of mechanical requirements are requested, the damaged fibres from the thermal recycling methods may still be viable. The section 3.1 about Thermal Recycling and the different studied methods all show promising results in recycling the resin matrix, especially solvolysis 3.1.4 and its ability to degrade the resin into monomers. However, the heating and high temperatures damage the fibres' properties and may strip them of their sizing. Applications with lower requirements regarding the reinforcement properties could be a viable route to successfully reuse the resin matrix and introduce it back into the production stream, for example as petrochemicals, and still find a use for the fibres. The thermal processes will however demand a cleaning step since it will leave char residue on the surfaces of the fibres. The solvolysis method will also require an additional step of reapplying size 2.1.2 to the fibres. From an energy demand aspect, the chemical and thermal recycling processes demand considerably higher amounts of energy. However, it is important to consider the additional processes that may come after mechanical recycling to get to the scale of monomers, like solvolysis. The conclusion is not always as clear as in Figure 3.6.

If a composite has been recycled many times before or is damaged to the point that there are no remaining recycle routes left for the composite material, the most suitable end-of-life application is feeding it into a cement kiln. As mentioned in 3.4.1, the process utilizes both the fibres' chemical composition and the resin matrix. As The European Composites Industry Association already stated this is the most effective way of utilizing a "dead" waste material. This is because the cement kiln uses both materials and energy. The ideal world where circularity is the norm would not prefer end-of-life applications, but rather reuse the materials again and again. Only when it has fully run its course, give it back to the planet. A fully circular life of the material. But the world is not ideal, certainly not the composite industry, which is why this thesis was created from the beginning. Making a small change is better than nothing when the planet stand on the brink of emptying our resources.

# 5

# Conclusion

This thesis concludes that there are several technically viable recycling and reuse methods available for GFRP. What is applicable to introduce will depend on several factors such as waste volume, contamination, current supply chains, cost, energy demand and more. These are topics for additional investigation. The most common changes to the material properties are loss in tensile strength and stiffness for the fibres. There can also occur removal of sizing and fibre length reduction. The results found several examples of viable methods and the eminent findings are as follows: recycling techniques for thermoplastics injection moulding thermoplastic waste 3.2.3, TPC-cycle recycling 3.3.1, Thermosaic or Thermoprime recycling for PIR, end-of-life thermoset GFRP waste in cement kilns 3.4.1 and, fluidized bed pyrolysis for contaminated waste 3.1.2. Commercialization of post-treatments and additional adjusting processes to regain mechanical properties have the potential to add even more variety to the pool of GFRP recycling methods.

There are still several questions and areas of needed improvement in the composite recycling research. Some of the main obstacles are too decide what to prioritize, recover intact fibres and resins versus commercial and economical feasibility. Should the market focus on one of the materials or both? Prioritizing the material of the composite with the biggest mass, the polymer, may be the way forward. There is also a need for the current waste supply chain to start accepting composites and mixed materials and raise the demand for new recycling technologies that can handle more than just post-industrial waste. It will be exiting to see what the mentioned methods will develop in the future. The prediction is, despite the obstacles, that the research is just starting to unravel techniques to deal with the growing waste from the composite industry. Since composites often replace heavier materials like metals, they already lower the material usage and often the environmental impact from the start. That is why it is important to develop a sustainable circular route and not go backwards and ban glass fibres. The aim for circular GFRP continues and glass fibers can definitely have a place in a circular economy in the future.

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# Appendix A

# Result Comparison Tables

Here are the original table figures included.

| ***Studied Methods*** | ***Resin rec.*** | ***Fibre rec.*** | ***Energy rec.*** | ***Fibre changes*** | ***Advantages*** | ***P*** | ***Drawbacks*** |
|---|---|---|---|---|---|---|---|
| Pyrolysis | No | Yes | Yes | Damage fibres | - Filler recovery<br>- Can use the produced gases and oils as fuel | / | - Char contamination on components<br>- Produces gas emissions<br>- Fibers degraded by high temperatures |
| Fluidized bed pyrolysis | No | Yes | Yes | Damage fibres | - Can treat contaminated and mixed materials<br>- No char residue | PCR | - Loss of tensile strength on fibres<br>- Lesser energy recovery compared to pyrolysis |
| Microwave assisted pyrolysis | No | Yes | Yes | Damage fibres | - Less harm to fibres because of lower temperature<br>- Degrades resin into monomers | / | - Char residue on fibres<br>- Only for microwave absorbing materials |
| Solvolysis | Yes | Yes | No | Loses sizing | - Resins return to monomer state and can easily be reused<br>- No char residue | / | - Expensive equipment<br>- High temperature and pressure conditions<br>- Possible hazardous solvent |
| Milling | Yes | Yes | No | Damage fibres | - Milled material can be used as filler<br>- Low energy cost | PCR | - Loss of mechanical properties for fibers, both length reduction and degradation<br>- Filler usage is limited by hard processability due to high viscosity |

*Figure A.1*

| ***Studied Methods*** | ***Resin rec.*** | ***Fibre rec.*** | ***Energy rec.*** | ***Fibre changes*** | ***Advantages*** | ***P*** | ***Drawbacks*** |
|---|---|---|---|---|---|---|---|
| High frequency | Yes | Yes | No | Poor adhesion | - Uses no chemicals, heating or water<br>- Breaks the weakest chemical bonds | PCR | - Start-up stage<br>- Need separation stage and cleaning off the polymer residue from the fibers |
| Thermosaic & Thermoprime | Yes | Yes | No | Stiffness reduc. | - Preserves mechanical properties. | PIR | - Only for thermoplastic manufacturing waste |
| ThermoPlastic Composite-Cycle Recycling | Yes | Yes | No | None | - Preserves mechanical properties.<br>- Retention of long fibres. | PIR | - One study show a slight degrading of the matrix, and a change in crystallinity. |
| Injection Molding Termoplastic Waste | Yes | Yes | No | - | - Equivalent mechanical properties for recycled fibres compared to virgin | / | - Only for thermoplastics<br>- Dissolution step requires volatile solvent<br>- Material recovery loss |
| Cement Kiln | No | No | Yes | - | - Can handle big volumes with no ultimate waste<br>- Utilize both material and energy | PCR | - One study reported only 67% material recovery and 33% energy recovery |

*Figure A.2*

| ***Studied Methods*** | ***Thermoplastics (TP) / Thermosets (TS)*** | ***Country*** | ***Commercially available*** | ***Associated companies*** |
|---|---|---|---|---|
| Pyrolysis | TS | Europe | Yes | - |
| Fluidized bed pyrolysis | TS | - | No | - |
| Microwave assisted pyrolysis | TS | - | No | - |
| Solvolysis | TS | Japan, USA | Yes | Innoveox<br>Adherent Technologies Inc<br>Panasonic Electric Works |
| Milling | TS / TP | - | Yes | - |
| High frequency recycling | TS / TP | Sweden | No | Librixer AB |
| Thermosaic & Thermoprime | TP | France | Yes | Cetim Cermat |
| ThermoPlastic Composite Cycle Recycling | TP | Netherlands | No | ThermoPlastic Composite Application Centre |
| Injection Molding Thermoplastic Waste | TP | - | - | - |
| Cement Kiln | TS | Germany, USA | Yes | Eco Wolf<br>ADM Isobloc |

*Figure A.3*